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There were several major results in this project. We determined the unusual kinetics of simple *heterogeneous* reactions in which the mobility or velocity of each reactant is drawn from a distribution. We also elucidated the rich variety of kinetics in reactions where particles move on ballistic trajectories. Next, we mapped out the kinetics of a simple catalysis model on a disordered surface. Enhanced reactivity was discovered which arises specifically from the surface disorder. Third, we found surprising behavior in the kinetics and spatial organization of reactants in two-species annihilation, $A + B \to 0$, when all particles move in the same direction. We then investigated the coarsening kinetics of a system of growing and competing domains which can be viewed as a toy model of war. Finally, we describe advances in a number of physical realizations of aggregation and annihilation.

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FINAL REPORT

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1. Foreword

There were several major results in this project. We determined the unusual kinetics of simple *heterogeneous* reactions in which the mobility or velocity of each reactant is drawn from a distribution. We also elucidated the rich variety of kinetics in reactions where particles move on ballistic trajectories. Next, we mapped out the kinetics of a simple catalysis model on a disordered surface. Enhanced reactivity was discovered which arises specifically from the surface disorder. Third, we found surprising behavior in the kinetics and spatial organization of reactants in two-species annihilation, $A + B \to 0$, when all particles move in the same direction. We then investigated the coarsening kinetics of a system of growing and competing domains which can be viewed as a toy model of war. Finally, we describe advances in a number of physical realizations of aggregation and annihilation. These topics are detailed in the following report.

1. Kinetics of Heterogeneous Reactions

(i) *Diffusion-Controlled Single-Species Annihilation:* We investigated the kinetics of diffusion-controlled *heterogeneous* single-species annihilation, $A_i + A_k \xrightarrow{K_{i,j}} 0$ [1]. Here A_i denotes species i with diffusivity D_i and the reaction rate matrix $K_{i,j}$ is a function of the diffusivities of species i and j. This accounts for the different reactivities in a system with distribution of particle masses.

For a discrete distribution of diffusivities, the rate equations predict that the least mobile species decays as t^{-1}, as in homogeneous annihilation. However, the more mobile species were found to each decay at a faster power-law rate, with an associated exponent that depends on the diffusivity ratio between the more mobile and the slowest species. For a continuous diffusivity distribution, the rate equations again show that the least mobile species predominates in the long time limit and that the decay of the more mobile species is described by non-universal power-law behavior. For an initial distribution of diffusivities with a power law tail, $P(D, t = 0) \sim D^{\mu}$, as $D \to 0$, the concentration and the average diffusion coefficient decay as $c(t) \sim t^{-\alpha}$ and $D(t) \sim t^{-\beta}$, respectively, with $\alpha = (2 + 2\mu)/(3 + 2\mu)$ and $\beta = 1 - \alpha = 1/(3 + 2\mu)$. These predictions are expected to apply for spatial dimension $d > 2$. For $d \leq 2$, we have adapted the Smoluchowski theory to account for the reaction kinetics. This theory yields predictions which predicts that the impurity species decays as a non-universal power-law in time and are in excellent agreement with numerical results from time-series expansions and Monte Carlo simulations in one dimension.

An additional intriguing feature is found in the *stationary impurity problem* in which there is a single stationary particle in a homogeneous reactive background. For this situation, the impurity survival probability decays as $t^{-3/8}$, while the background density decays as $t^{-1/2}$. This is one of the first examples of the newly-discovered problem of *persistence* in reaction kinetics [2], in which the probability that a given point in space is not affected by the reaction decays in time with a new independent exponent. Our Smoluchowski theory provide a conceptually simple and quantitatively accurate description of this result.

(ii) *Ballistic Reactions:* The kinetics of the annihilation process, $A + A \to 0$, with ballistic particle motion was investigated when the distribution of particle velocities is *discrete* [3-5]. This discreteness is the source of many intriguing phenomena. In the mean field limit, the densities of different velocity species decay in time with different power law rates for many initial conditions. For a one-dimensional symmetric system containing particles with velocity 0 and ± 1, there is a particular initial state for which the concentrations of all three species decay as $t^{-2/3}$ [4,5]. For the case of a fast "impurity" in a symmetric background of + and − particles, the impurity survival probability decays as $\exp(-\text{const.} \times \ln^2 t)$. In a symmetric 4-velocity system in which there are particles with velocities $\pm v_1$ and $\pm v_2$, there again is a special initial condition where the two species decay at the same rate, $t^{-\alpha}$, with $\alpha \cong 0.72$. Efficient algorithms were developed to perform the large-scale simulations necessary to observe these unusual phenomena clearly.

(iii) *Traffic Flow Model:* We investigated a simple aggregation model that mimics the clustering of traffic on a one-lane roadway [6]. While many models of traffic flow have been introduced [7-9], the clustering that arises for heterogeneous traffic on a strictly one-dimensional roadway with no passing was not previously examined. Our model is an infinite one-dimensional roadway in which each "car" moves ballistically at its initial velocity until it overtakes the preceding car or cluster. After this encounter, the incident car assumes the velocity of the cluster which it has just joined. The properties of the initial distribution of velocities in the small velocity limit control the long-time properties of the aggregation process associated with clustering of cars. An exact analysis, based on the statistics of extremes, yields a formally exact solution for the process. For an initial velocity distribution with a power-law tail at small velocities, $P_0(v) \sim v^\mu$ as $v \to 0$, a simple scaling argument shows that the average cluster size grows as $n \sim t^{(\mu+1)/(\mu+2)}$ and that the average velocity decays as $v \sim t^{-1/(\mu+2)}$ as $t \to \infty$. (We may set the velocity of the "slowest" car to zero with loss of generality.) We also derived a complete analytical solution for the survival probability of a single car and an asymptotically exact expression for the joint mass-velocity distribution function. Our detailed results agree with those from simple-minded scaling arguments. We also investigated the evolution of an initially empty system with a steady input of cars. We found that this system exhibits a steady state for the case where the velocity distribution of input cars is $P(v) \sim v^\mu$ with $\mu > 0$. However, in the complementary case of $\mu < 0$, the system remains continuously evolving with both the concentration and the velocity exhibiting an algebraic temporal behavior.

(iv) *Multiscale phenomena:* We investigated the kinetics of single-species annihilation, $A + A \to 0$, in which each particle has a fixed velocity, which may be either $\pm v$ with equal probability, and also a finite diffusivity [10]. The intriguing feature is that even though the density decays as $t^{-1/2}$ for both the diffusion-controlled and convection-controlled reactions, the interplay between convection and diffusion leads to a decay of the density which is proportional to $t^{-3/4}$. Further, at long times, the reactants organize into domains of right- and left-moving particles, with the typical distance between particles in a single domain growing as $t^{3/4}$, and the distance between domains growing as t. The distribution functions between nearest-neighbor distances exhibits strong identity-dependent features. There is a very slow approach to this multiscale state as a function of time. Many of these results can be obtained by simple dimensional arguements [10].

2. Catalysis on a Disordered Surface

We considered the effects of quenched surface imperfections on the reaction kinetics of the idealized monomer-monomer model [11]. Here A and B particles impinge upon a surface, with respective rates k_A and k_B, and adsorb onto vacant sites. Nearest-neighbor adsorbed AB pairs react and desorb with rate k_r, leaving behind two vacancies. When all surface sites have the same adsorption characteristics, this model has simple kinetics: for $k_A \neq k_B$, the adsorption imbalance leads to the surface becoming saturated by the majority species and the reaction stops, while for $k_A = k_B$, there is a fluctuation-induced coarsening of the surface into growing A and B adsorbed islands. When islands become of the order of the size of the surface, there is no longer available AB reactive interface and the reaction stops. This saturation time is proportional to L^2 for an L-site chain and proportional to $L^2 \ln L$ for an $L \times L$ surface. Thus there is no reactive steady state on a homogeneous surface [11].

Our primary result is that surface disorder can create both a reactive steady state, as well as new "phases" with anomalously slow approach to saturation [12]. Further, a rich variety of kinetic behavior occurs which stems from the interplay between the surface disorder and the relative adsorption rates of the two species. Our model involves the following steps

$$A + S \xrightarrow{k_{A,S}} A_S$$

$$B + S \xrightarrow{k_{B,S}} B_S$$

$$A_S + B_S \xrightarrow{k_r} AB \uparrow + 2S.$$

Here the adsorption rates of each species, $k_{A,S}$ and $k_{B,S}$, depends on the surface site, as well as on the incident particle fluxes. We considered dichotomous disorder for the surface sites: one type, $+$, with concentration c_+, prefers to adsorb A, while the complementary $-$ sites, with concentration $c_- = 1 - c_+$, prefer to adsorb B. If the relative fluxes of A and B are p and $q = 1 - p$, then the disorder is defined by the rule that a vacant $+$ site becomes occupied by an A with probability $p + \epsilon$ and by a B with probability $q - \epsilon$, whereas a vacant $-$ site becomes occupied by an A with probability $p - \epsilon$ and by a B with probability $q + \epsilon$. The strength of the adsorptive disorder is thus quantified by the parameter ϵ.

In the mean-field limit we found a new disorder-stabilized reactive steady state over a finite region of the (p, ϵ) phase diagram in which $x = x_\infty(p, \epsilon, c_+)$ and $y = y_\infty(p, \epsilon, c_+)$. Here $x \equiv n_A^+/N$ and $y \equiv n_A^-/N$ as the respective concentrations of adsorbed A on the $+$ and $-$ sites, where N is the total number of surface sites. The respective concentrations of B on the $+$ and $-$ sites are then $c_+ - x$ and $c_- - y$.

In one dimension, new kinetics regimes arise, whose qualitative characters are independent of the precise nature of the surface disorder. For $p + \epsilon < 1/2$, B is preferentially adsorbed on both the $+$ and $-$ sites and the concentration of adsorbed A, $C_A(t)$, decays exponentially in time. This range defines the **B**-phase of the disordered system in which the minority species vanishes in time as $C_A(t) \sim C_{AB}(t) \sim \exp(-at)$ with $a = a(p, \epsilon, c_+)$. On the other hand, for $1/4 < p < 1/2$ and $1/2 - p < \epsilon < p$ in the phase diagram, the average adsorption rate globally favors B but $+$ sites adsorb A with a probability greater than $1/2$. This dichotomy now leads to a power law decay of $C_A(t) \sim t^{-\beta}$, with a non-universal

exponent $\beta(p, \epsilon, c_+)$. This region of the phase diagram corresponds to a new state, denoted the **b**-phase, in which there is B saturation, but with slow power-law kinetics.

For the physical case of a two-dimensional disordered surface, five phases occur, including a new disorder-stabilized reactive steady state **R**. As in one dimension, it may established rigorously that in the **A** phase, where A is preferentially adsorbed on both the + and − sites, A-saturation is approached exponentially in time. This conclusion is independent of the values of c_+ and c_-. In numerical simulations on a square lattice of 10^6 sites, we have also found **a** and **b** phases for the symmetric case $c_+ = c_- = 1/2$. In these regions the minority species decays as a power law in time, $C_{\text{minority}}(t) \sim t^{-\beta_m}$, with a non-universal exponent $\beta_m = \beta_m(p, \epsilon, c_+)$. Finally, in the reactive steady state (**R**) the densities approach nonzero values exponentially in time. This result could have wide application, as previous investigations have observed a reactive steady state on in the case of a more complicated stoichiometry such as the monomer-dimer reaction [13] to describe the oxidation of CO on a metal surface

3. Two-Species Annihilation with Driven Motion

We studied the kinetics and related spatial organization effects for two-species annihilation in one dimension, $A + B \to 0$, when all particles undergo strictly biased motion in the same direction and with an excluded volume repulsion between same species particles [14,15]. That is, each particle can attempt a move only to the right and the attempt is successful only if the particle lands on an unoccupied site. If the particle lands on a site already occupied by a member of the same species, the move attempt is rejected and the initial particle remains at its starting position. However, if the particle lands on a site which is occupied by a member of the opposite species, then annihilation occurs and both particles disappear. Naively, one would expect by making a Galilean transformation to the "rest" frame, that the effect of the bias would be irrelevant. However, numerical simulations [16] showed that the density in this system decays as $t^{-1/3}$, compared to the anticipated $t^{-1/4}$ density decay in the system with isotropic diffusion.

We suggested a simple explanation for this surprising result based on analysis of the Burgers equation [17] with boundary conditions appropriate for describing the domain structure of reactants. Related properties associated with the asymptotic distribution of reactants can also be accounted for within this Burgers equation description. Our approach is based on first elucidating the dynamics of a single AB interface when the two species are initially separated. A continuum description in terms of the inviscid Burgers equation, together with continuity of particle currents, shows that the velocity of the interface can be determined in terms of the relative densities of the two adjoining species. This information is used to infer that when the initial condition is homogeneous and random, a typical single-species domain grows as $t^{2/3}$. This result, together with the fact that the density inside a domain is simply related to initial density fluctuations, leads to the $t^{-1/3}$ decay of the density. We also performed numerical simulations of the reaction, focusing, in particular, on the density decay exponent, the domain length growth exponent, and the behavior of the scaled domain density profile. In addition to confirming our theoretical results, we also find that the average density profile of a domain exhibits a shock wave, whose scaling properties involve a new independent length scale.

4. Coarsening of Domains at "War"

A model of growing and annihilating domains that mimics some essential features of human history was investigated [18]. Continuously growing civilizations are represented by domains, which engage in "war" whenever two domains meet. This model bears some resemblance to models of breath figure growth [19], where droplets grow in a supersaturated environment and coalesce upon meeting. Here we investigate the situation where the droplets destructively complete upon contact by undergoing a "war". In a war, the smaller domain is annihilated, while the larger domain shrinks by a fraction ϵ of the casualties of the loser. Thus ϵ quantifies the fairness of the war, with $\epsilon = 1$ corresponding to a fair war, with equal casualties on both sides, and $\epsilon = 0$ corresponding to a completely unfair war, where the winner suffers no casualties. When new civilizations are continuously being born into this warring world, two different long-time behaviors arise as a function of ϵ. When $1/2 < \epsilon \leq 1$ (relatively fair wars), a steady state arises, which is characterized by egalitarian competition between domains of comparable size. For the limiting case of $\epsilon = 1$, the steady state is characterized by domains whose age is typically much larger than their size. When $0 \leq \epsilon < 1/2$ (relatively unfair wars), "superpowers" ultimately dominate. This coarsening process is characterized by power-law temporal behavior, with non-universal ϵ-dependent exponents. These features are captured by a deterministic self-similar model, for which the characteristic exponents can be computed easily. The transition point $\epsilon = \epsilon_c = 1/2$ is characterized by anomalously slow coarsening.

5. Kinetics of Aggregation and Annihilation

(i) *Charged particle annihilation:* The annihilation kinetics of a two-component plasma-like system of positive and negative charges was considered [20]. Similar charges repel and opposite charges attract with a force which decays with distance as $F(r) \sim r^{-\lambda}$. When two oppositely charged particles meet, they irreversibly annihilate. In the long-time limit, the reactants segregate into single-species domains, and the late time kinetics is determined by the motion of domain interfaces. For sufficiently large λ (short-range interaction), diffusive particle motion prevails over the force-induced drift, leading to familiar diffusion-limited aggregation. For smaller λ values, the interparticle interaction becomes essential, resulting in non-universal algebraic decay of the particle concentrations. For even smaller λ, the typical force acting on a particle becomes system size-dependent and the concentration decays faster than any power law in time.

(ii) *Aggregation with evaporation and condensation:* We investigated the kinetics of aggregation [21] which is augmented by: (a) evaporation of monomers from clusters – "dry" aggregation, or (b) continuous cluster growth or condensation – "damp" aggregation. The rate equations for these two processes were analyzed using both exact and asymptotic methods. In dry aggregation, the competition between evaporation and aggregation leads to several asymptotic outcomes. When the evaporation is weak, the kinetics is similar to that of aggregation with no evaporation, while a steady state is quickly reached in the opposite case. At a critical evaporation rate, a steady state is slowly reached in which the cluster mass distribution decays as $k^{-5/2}$, where k is the mass, while the typical cluster

mass, or upper cutoff in the mass distribution, grows with time as $t^{2/3}$. For damp aggregation, the typical size typically grows faster than linearly in time, with the quantitative nature of this growth dependent on system details.

(iii) *Multiple conservation laws:* Aggregation processes with an arbitrary number of conserved quantities were investigated [22]. Within mean-field theory, the exact size distribution was obtained. The asymptotic form of this solution exhibits nontrivial "double" scaling in which an additional diffusion-like mass scale arises from the multiple conservation laws. Our theory was applied to ballistic aggregation with mass and momentum conserving collisions and to diffusive aggregation with multiple species.

(iv) *Diffusive capture process:* The survival probability, $S_N(t)$, of a diffusing "prey" particle in the proximity of N diffusing "predatory" particles was investigated [23]. In one dimension, surprising kinetic behavior is observed when predators are all to one side of the prey. In this case, the survival probability decays as a non-universal power law, $S_N(t) \propto t^{-\beta_N}$, with the decay exponent β_N proportional to $\ln N$. When $N \to \infty$, the prey survival probability exhibits a log-normal decay, $S_\infty(t) \propto \exp(-\ln^2 t)$. This last result substantially extends previous mathematical work on this subject [24,25].

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